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Specification and Drawings, as originally filed, with Application for Patent Serial No: **2,273,761**, on June 9, 1999, by **POLYVALOR S.E.C.**, assignee of Danilo Klavana, Jitka Kirchnerova, Jamal Chouki and Christophe Guy, for "Apparatus and Process for Catalytic Gas Involving Reactions Which are Exothermic".

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July 7, 2000

Date

Title of invention: **Apparatus and Process for Catalytic Gas Involving Reactions which are Exothermic.** (Apparaillage et procedé pour les reactions catalytiques exothermiques.)

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Description of invention:

This invention pertains to the catalytic gas involving reactions which are exothermic. In particular, the invention relates to a new (improved) and economical apparatus to carry out such reactions (processes). The new apparatus consists of a two compartment concentrical self regulating (autocyclic) reactor, which may be operated in any convenient position. The reaction space of the reactor is filed with suitable catalytic material. The gas stream enters the outer compartment and flows across the catalytic bed of the outer compartment to the end of the reactor, where it is forced to enter the inner central compartment. The inner central compartment is also filled with a catalytic material across which the gases now flow in a direction opposite to that of the flow in the outer compartment. Different types of catalytic materials may be employed, along with an inert material, depending on the facility of the reaction, its heat content and on the size of the reactor. The rate and direction of the reaction front displacement depend on the volumetric heat balance. If the front moves up to the exit part of the inner reactor compartment which is thermally communicating with the entrance part of the reactor via fins its heat re-ignites the incoming gas reaction mixture and a new cycle begins. The schema of the apparatus in shown in Figure 1.

The apparatus and the process are especially suitable for highly exothermic reactions of total oxidation in air of various combustible gases present at relatively low concentrations. These reactions are exploited either for heating (using natural gas, propane or other suitable gaseous fuel), or for destruction of a variety of undesirable combustible vapors and gases in industrial effluents. The apparatus and the process may also be used for the production of sulfur trioxide.

Background

Catalytic combustion of lean fuel/air mixtures, whether it is for heat generation, or for cleaning gas streams of combustible contaminants (VOC abatement), is the safe, environmentally most preferred alternative to traditional flame combustion(or incineration), which is a source of noxious nitrogen oxides.

Operation of catalytic highly exothermic reactions involving total oxidation in classical fixed (stationary) bed reactors becomes very difficult to control when the concentration of the fuel as well as the temperature of the feed mixture are variable, because these conditions cause the reaction front to creep along the reactor axis. To remedy these and other problems such as creation of hot spots and overheating the catalytic bed variety of solutions have been proposed and patented. Elegant solution to the creep of the reaction front in the case of gas streams containing relatively low concentrations of fuel is the invention of Houdry, which consists of reverse-flow reactor comprising in addition to a catalytic bed a bed of inert material serving as a heat recuperator. Originally proposed for treatment of gas streams the reverse-flow (or cyclic feed) type of fixed-bed reactor (Houdry, 1960) has been modified by several inventors with a variety of improvements such as means of purging of regenarative incinerators of untreated gas stream and reintroducing it back to the system (Houston, 1975 ; Wojciechowski, 1991 ; Thunstrom, 1993) and for different specific applications (Matros et al., 1984 ; Grozev et al., 1997). The latest improvement of the reverse-flow regenerative type systems consists of providing means for controlling the temperature of the bed, by introducing additional chambers allowing the dilution of the reaction mixture (Chaouki et al., 1997). While all these inventions provide a number of advantages over a classical fixed bed reactors, these improved reverse-flow systems are relatively complex and bulky, requiring use of additional valves increasing thereby the investment, operation and maintenance cost. Several of these disadvantages are avoided when using the apparatus of this invention.

The advantages of the present invention will become clearly apparent with the help of drawings illustrating the general features of the apparatus (Fig. 1), schema of the laboratory experimental unit used to provide experimental demonstration of the operation (Fig.2), and the temperature profiles and reaction front displacement for two examples in which propane (Fig. 3-8) or methane (Fig. 9-13) were used as the fuel for producing hot air.

Novelty of the invention

A new stationary-bed reactor configuration which takes advantage of the heat release of exothermic reactions in a way to assure a self-regulating reactor operation.

Advantages

In comparison with current state of art reverse-flow (cyclic-feed) reactors the apparatus of this invention is characterized by simplicity, compactness, low maintenance, higher versatility.

Applications of the invention:

Wide range of applications can be envisaged. These applications involve exothermic catalytic reactions involving gas streams of variable relatively low reactant concentrations and variable inlet temperatures. Such reactions include:

catalytic combustion of lean fuel/air mixtures to produce hot air
catalytic cleaning of gas streams (effluents) containing combustible gases or vapors
production of sulfur trioxide from relatively low concentrated sulfur dioxide containing gases

Prove of concept:

The function of the apparatus of the invention and the feasibility of its use as a heat generator has been tested experimentally using both methane or propane as a fuel, as documented by data in Figures 3 to 13 and as described in the following Examples 1 and 2.

Potential commercialization:

Due to its versatility and potentially large area of applications, such as heating systems for industrial, commercial and residential use, catalytic cleaning (methane removal) of ambient air in mines and other closed spaces naturally contaminated by emanating gases, or VOC abatement to name only the most obvious, the invention has a great potential. Its commercialization should be pursued.

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__Keywords:__ Reactor, fixed bed, stationary bed, reverse-flow, regenerative ; catalytic combustion, heating ; catalytic air cleaning ; catalytic incineration of combustible, organic vapors, gases ; exothermic oxidation reactions.

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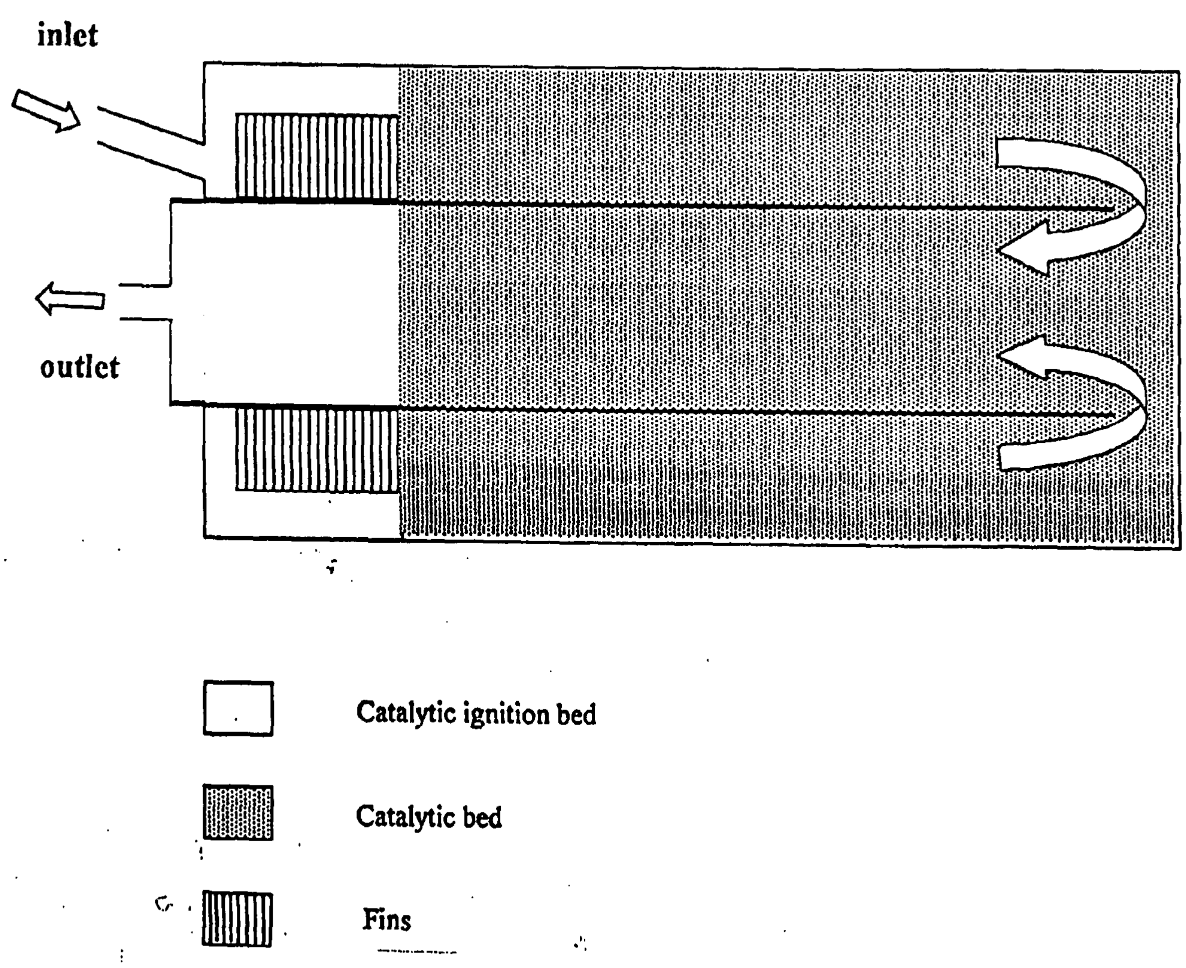

Fig. 1 : Schema of the apparatus according to the present invention

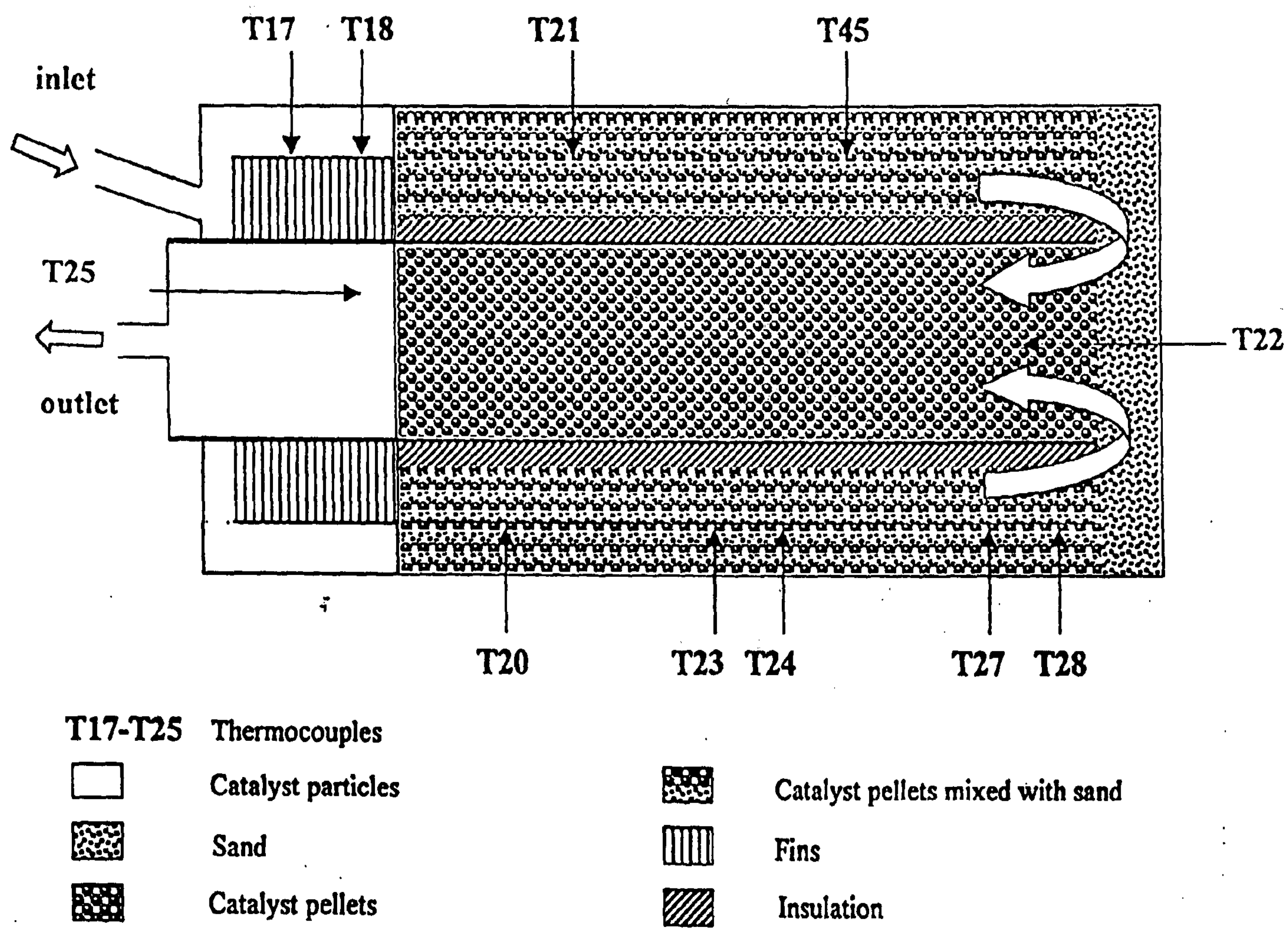

Fig. 2 : Schema of the experimental self regulating autocyclic reactor used for the demonstration

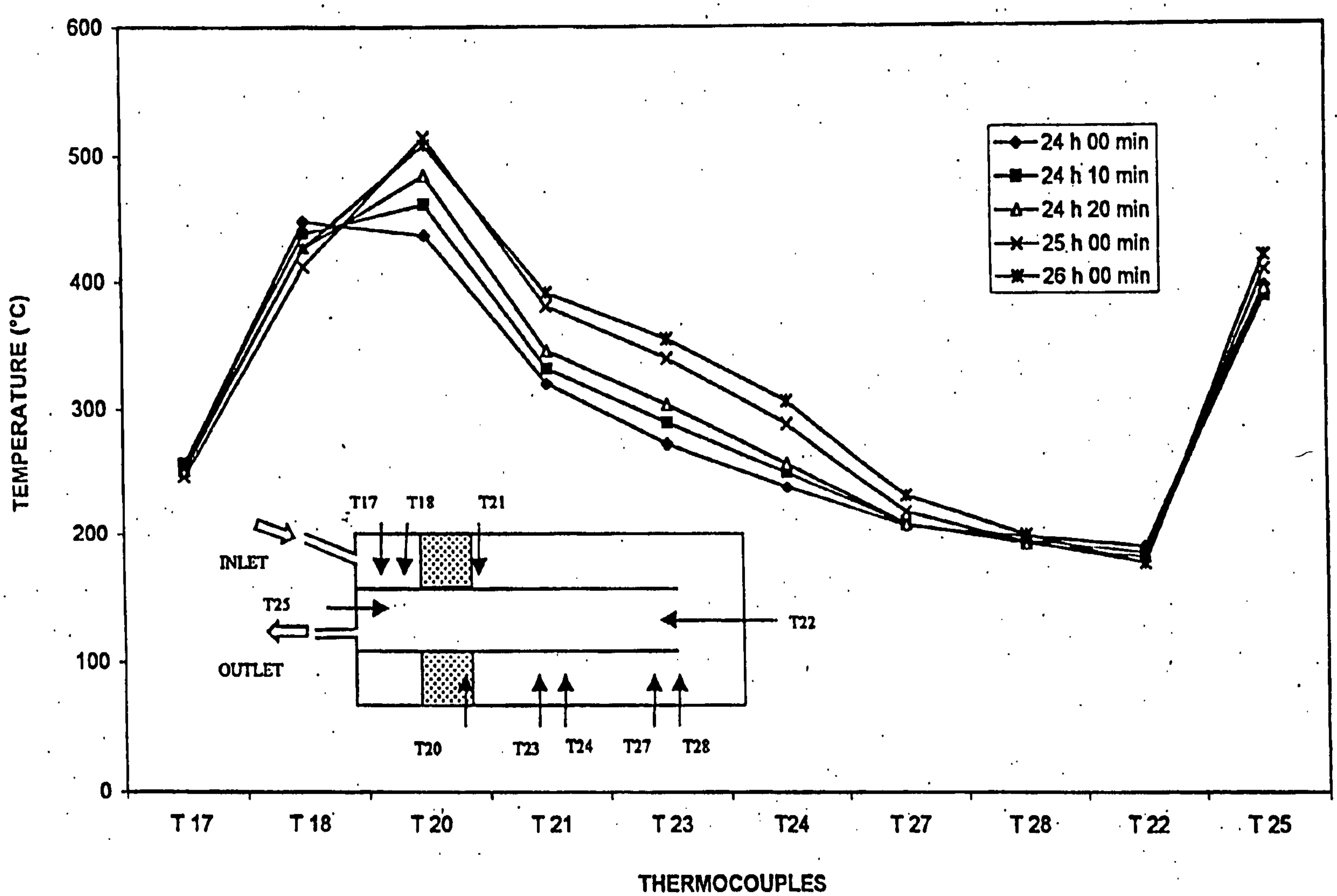

Fig. 3 : Temperature pr file and the position of the reaction front in the experimental self regulating react r f r 0.88% propane combustion, total flowrate 16.95 L/min (air flowrate : 16.8 L/min, propane flowrate : 0.15 L/min, propane concentration : 0.88 %).

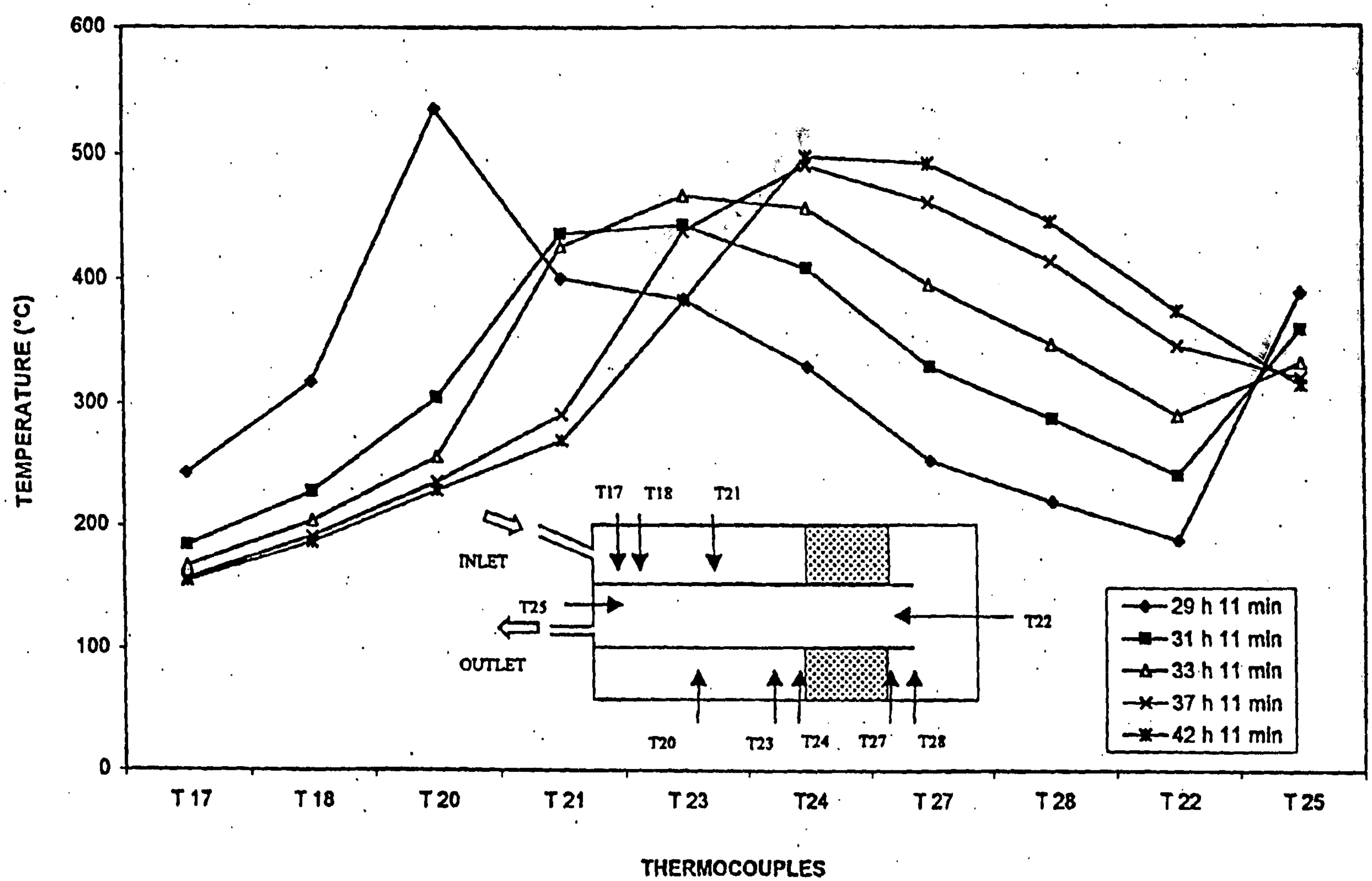

Fig. 4 : Temperature profile and the position of the reaction front in the experimental self regulating reactor for 0.46% propane combustion, total flowrate 22.25 L/min (air flowrate : 22.1 L/min, propane flowrat : 0.15 L/min, propane concentrati n : 0.67%).

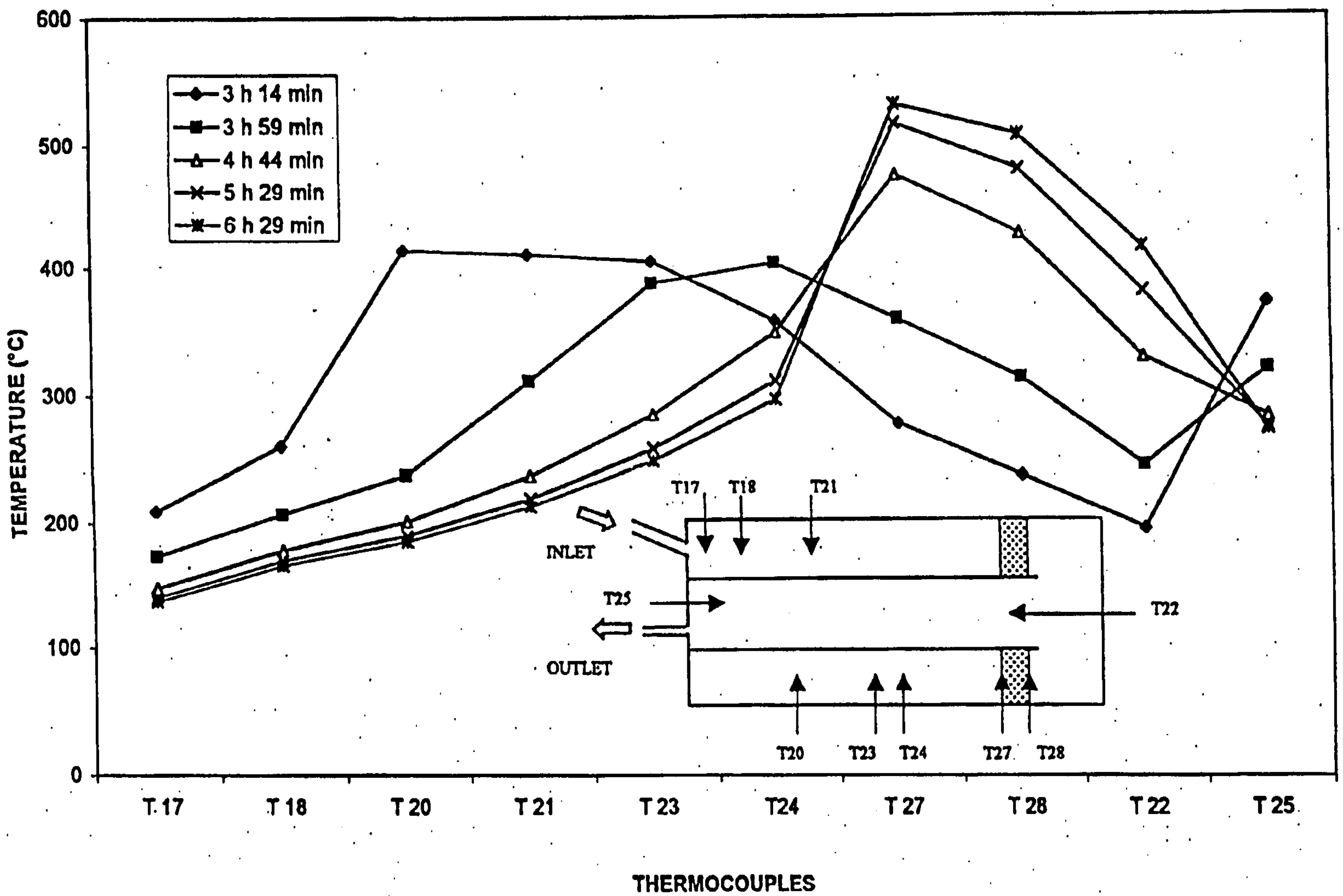

Fig. 5 : Temperature profile and the position f the reaction front in the experimental self regulating reactor for 0.55% propane combustion, total flowrate 27.55 L/min (air flowrate : 27.4 L/min, propane flowrate : 0.15 L/min, pr pane c ncentration : 0.55%).

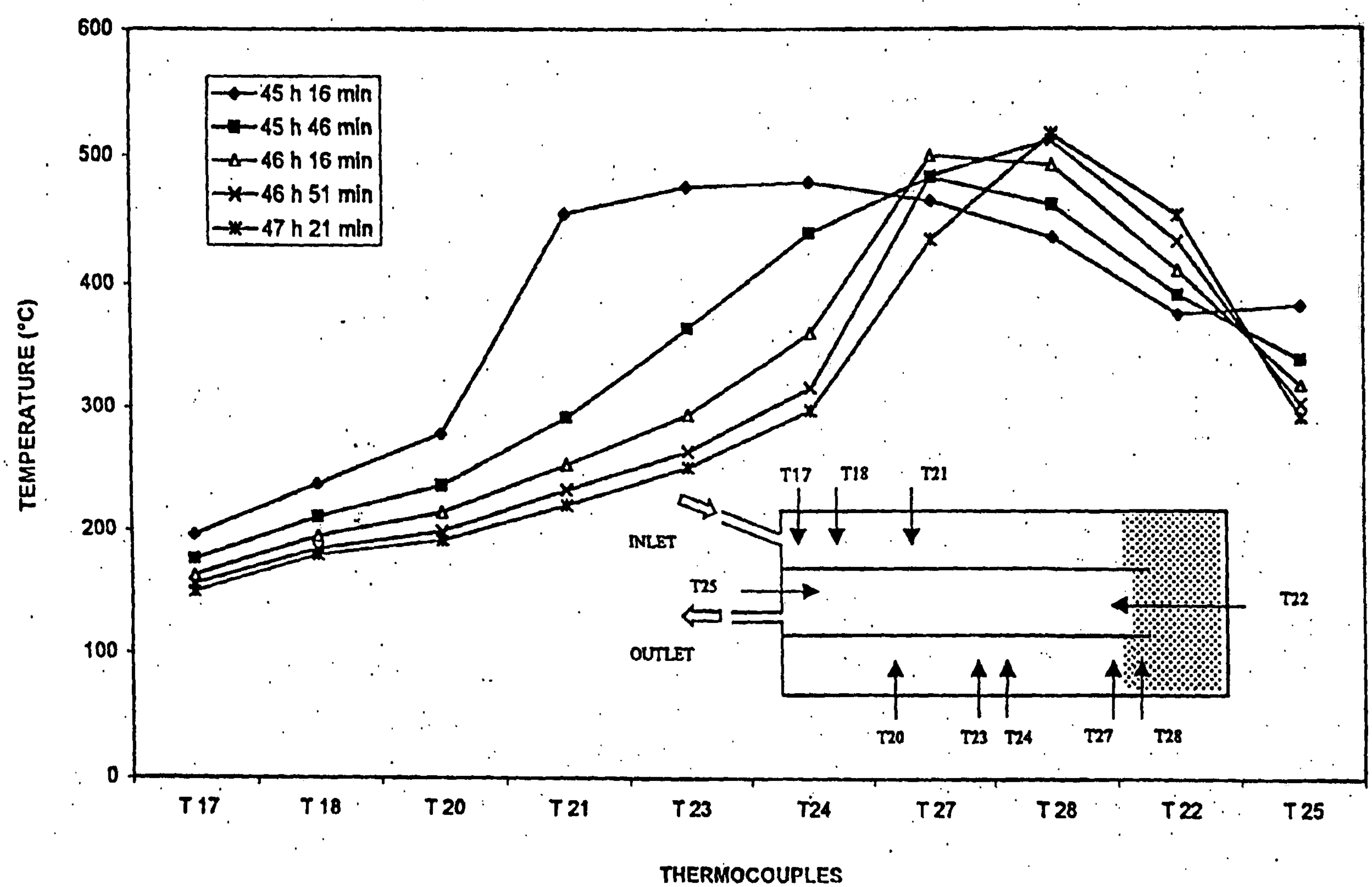

Fig. 6 : Temperature profile and the position f the reaction front in the experimental self regulating react r for 0.46% propane combusti n, total flowrate 32.85 L/min (air flowrate : 32.7 L/min, propan flowrate : 0.15 L/min, propane concentration : 0.46 %).

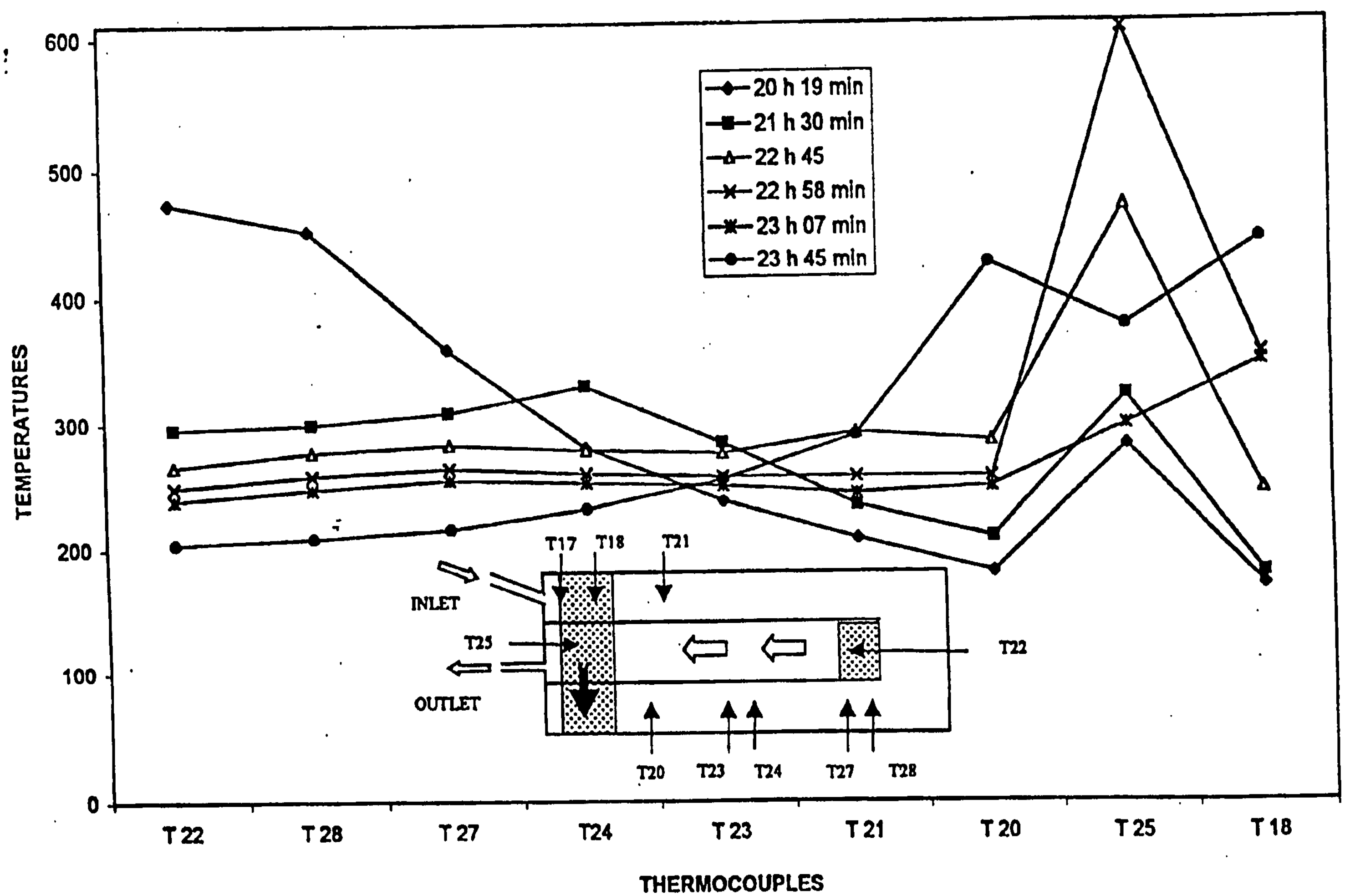

Fig. 7 : Temperature profil representing the displacement of the reacti n front and its re-ignition at the react r inlet f r the propane combusti n.

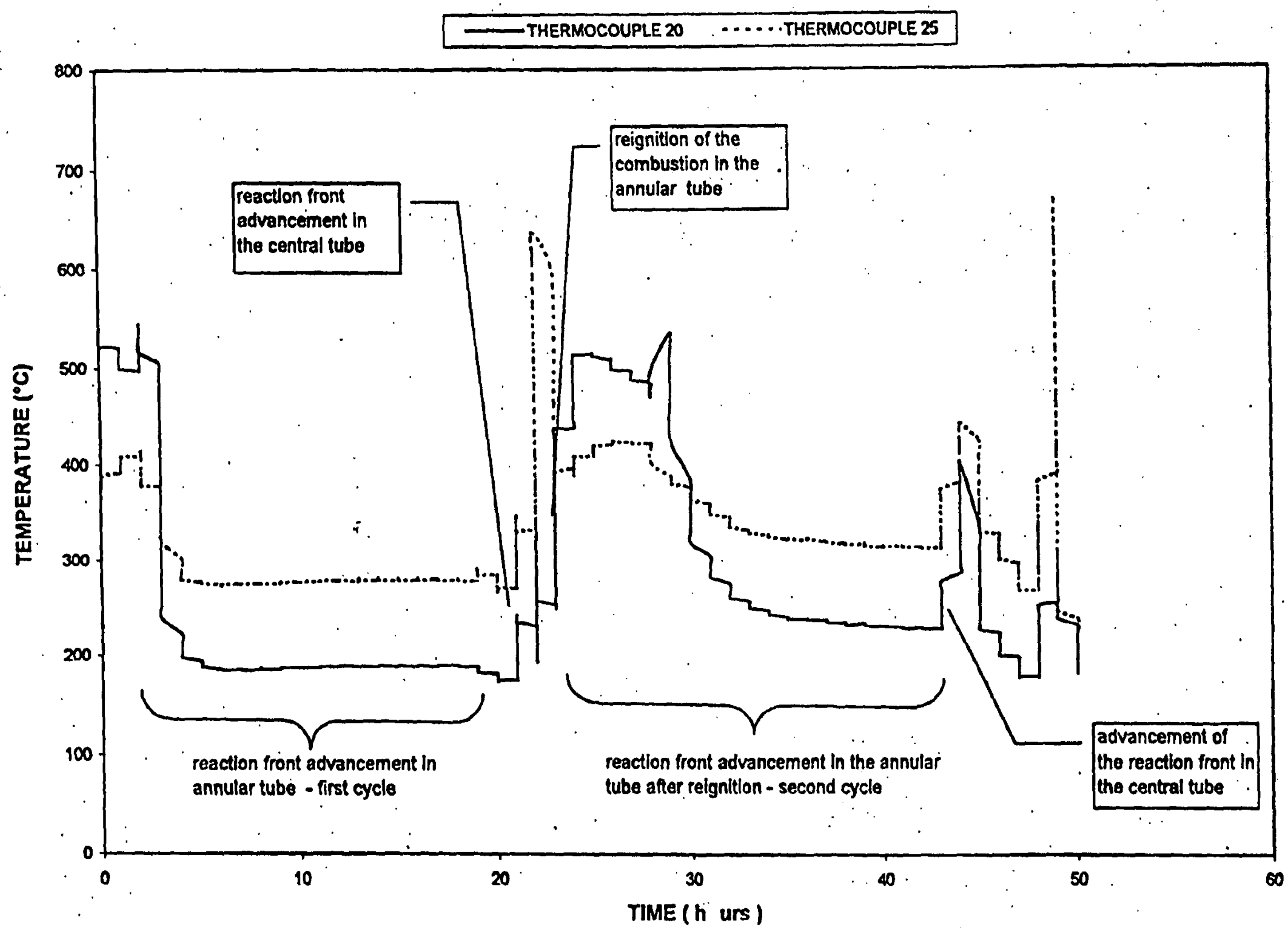

Fig. 8 : Temperature profiles representing autocyclic operation of the experimental self regulating reactor during propane combustion.

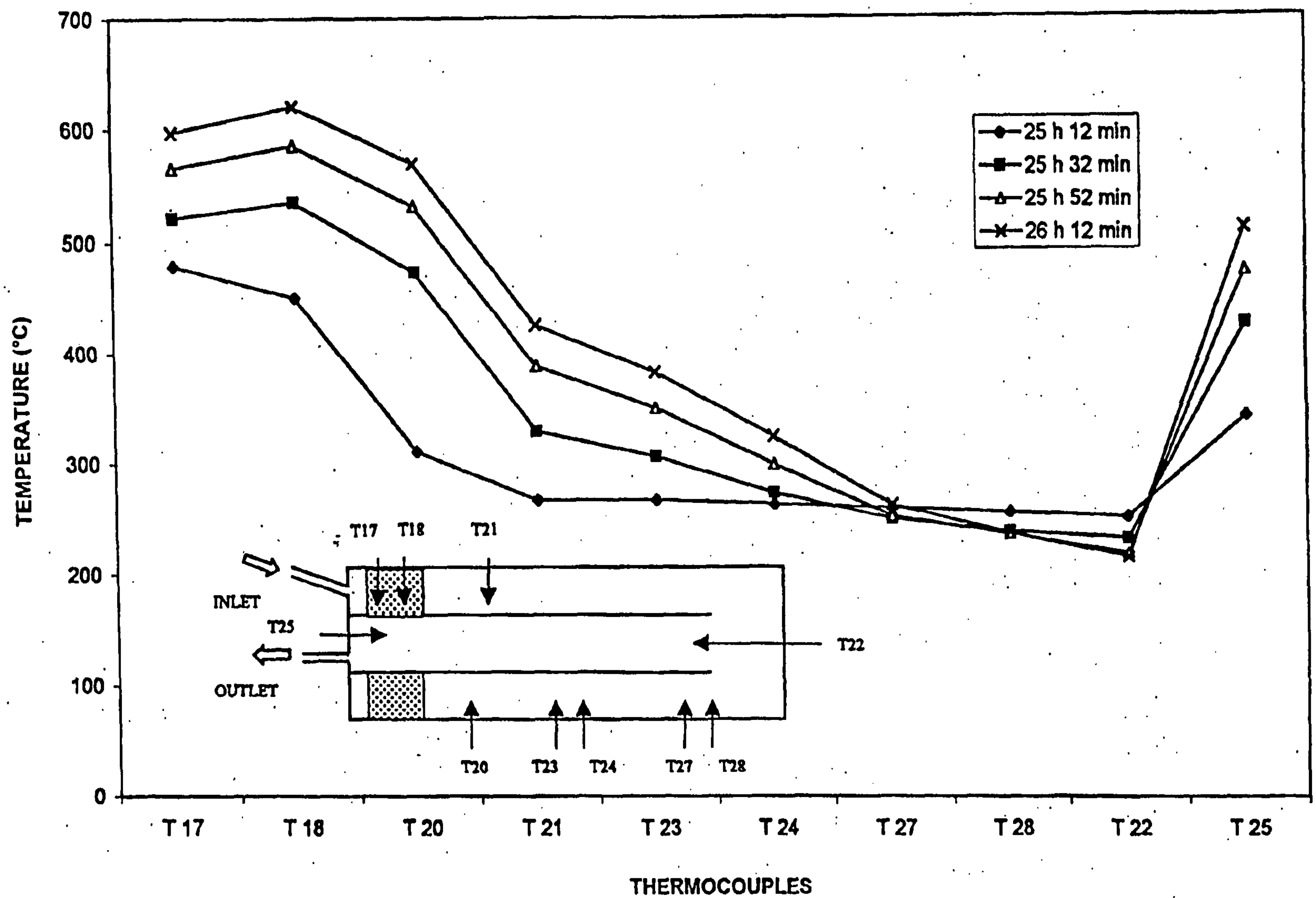

Fig. 9 : Temperature profil and the position of the reaction front in the experimental self regulating reactor for 5% methane combustion, total flowrate 20.52 L/min (air flowrate : 19,5 L/min, methane flowrate : 1,02 L/min, methane concentration : 5 %).

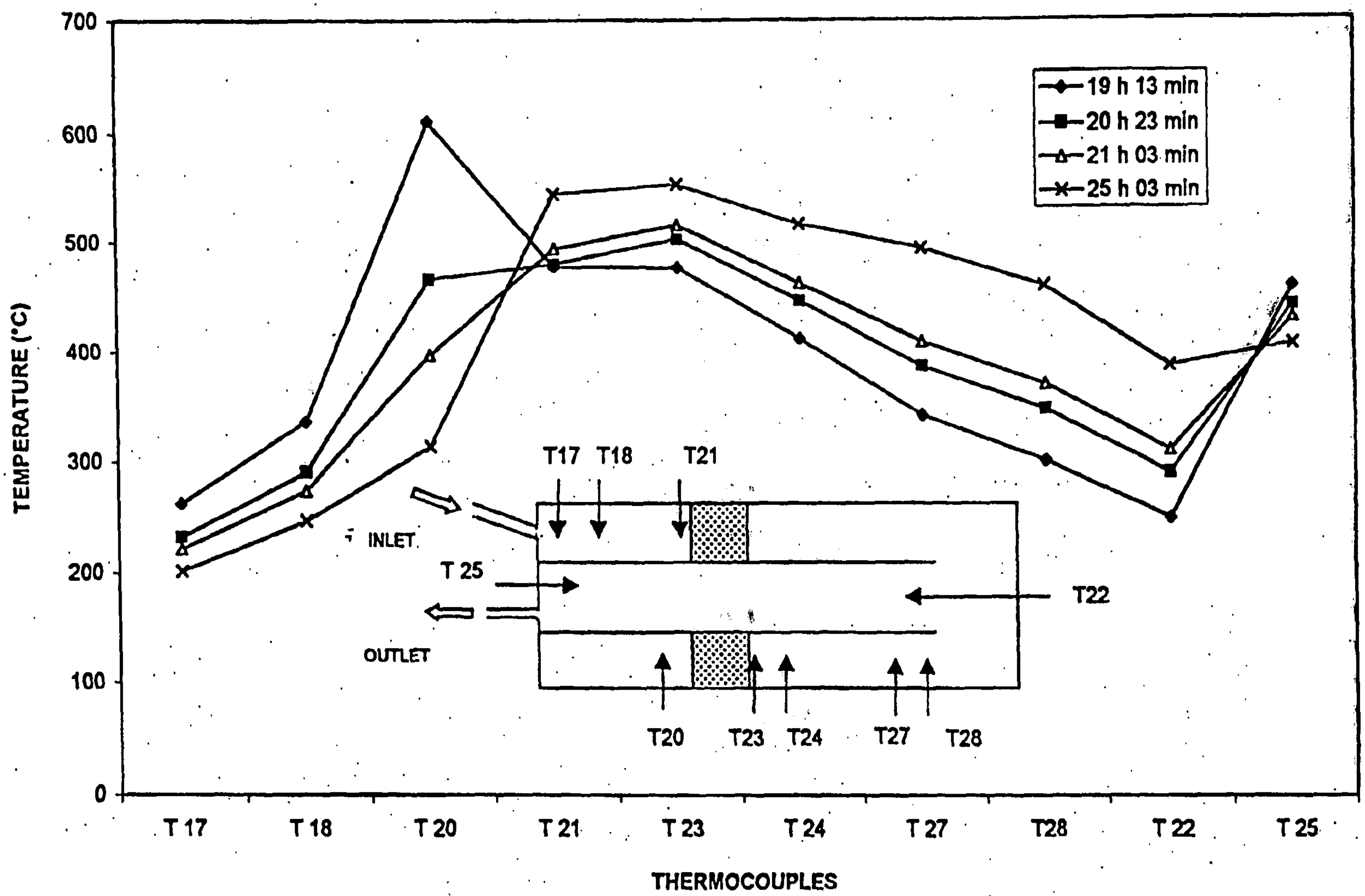

Fig. 10 : Temperature profile and the position f the reaction front in the experimental self regulating reactor for 1.98% methane combustion, total flowrate 25.2 L/min (air flowrate : 24.7 L/min, methane flowrate : 0.5 L/min, methane concentration : 1.98 %).

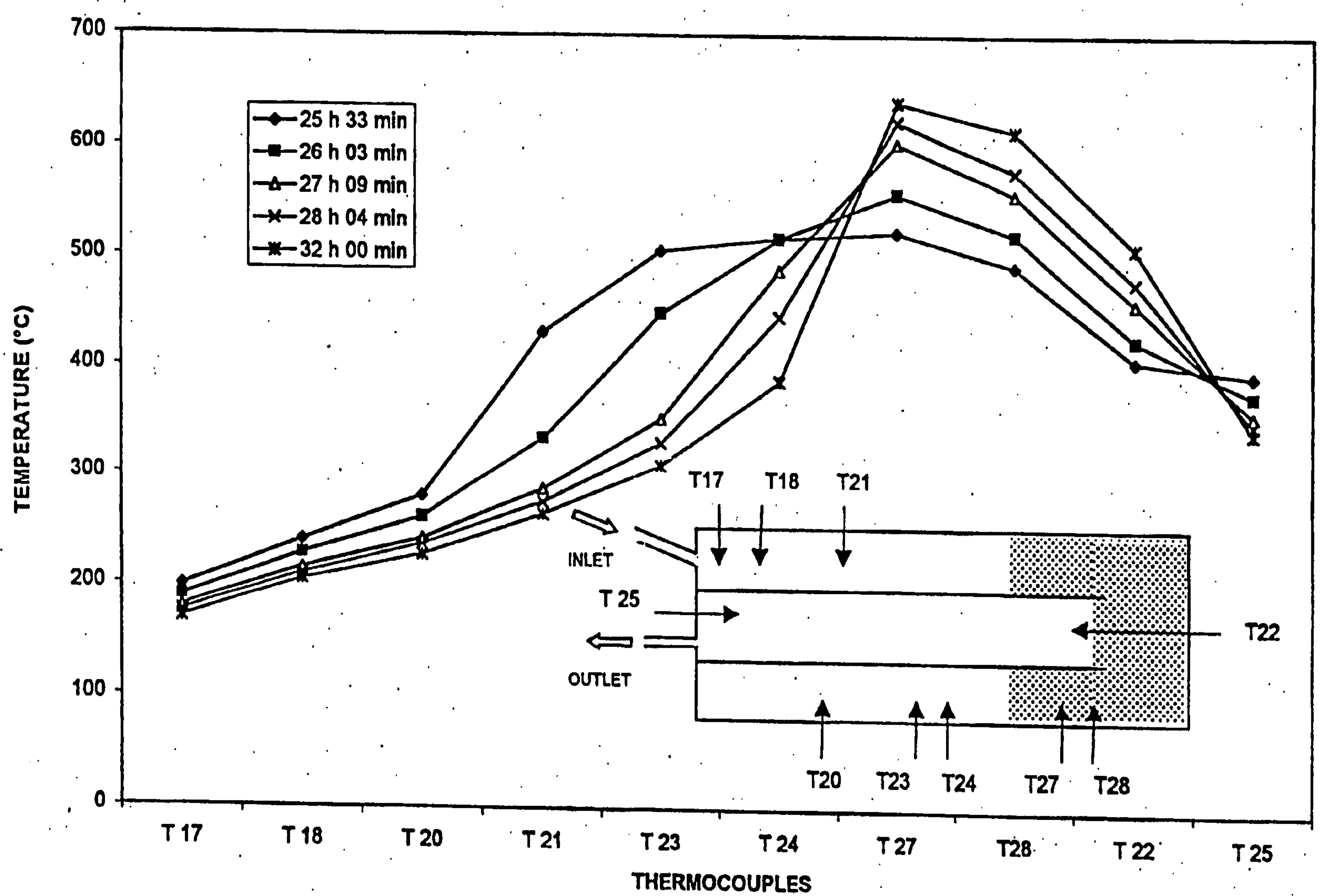

Fig. 11 : Temperature profile and the position of the reaction front in the experimental self regulating react r for 1.4% methane combustion, t tal fl wrate 30.5 L/min (air fl wrate : 30 L/min, methan flowrate : 0.5 L/min, methane c ncentration : 1.64 %).

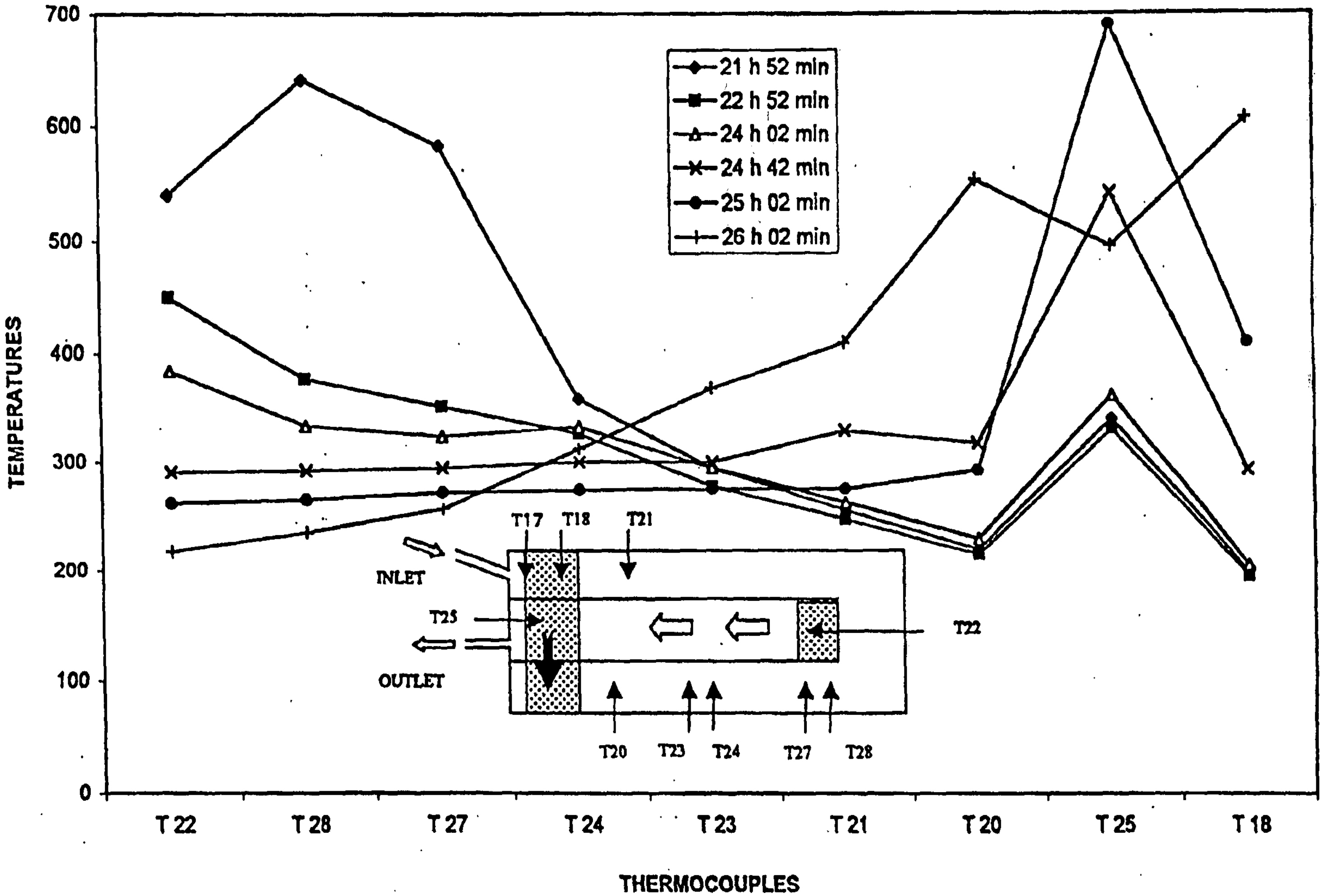

Fig. 12 : Temperature profile representing the displacement f the reaction front and its re-ignition at the react r inlet f r the methane combusti n.

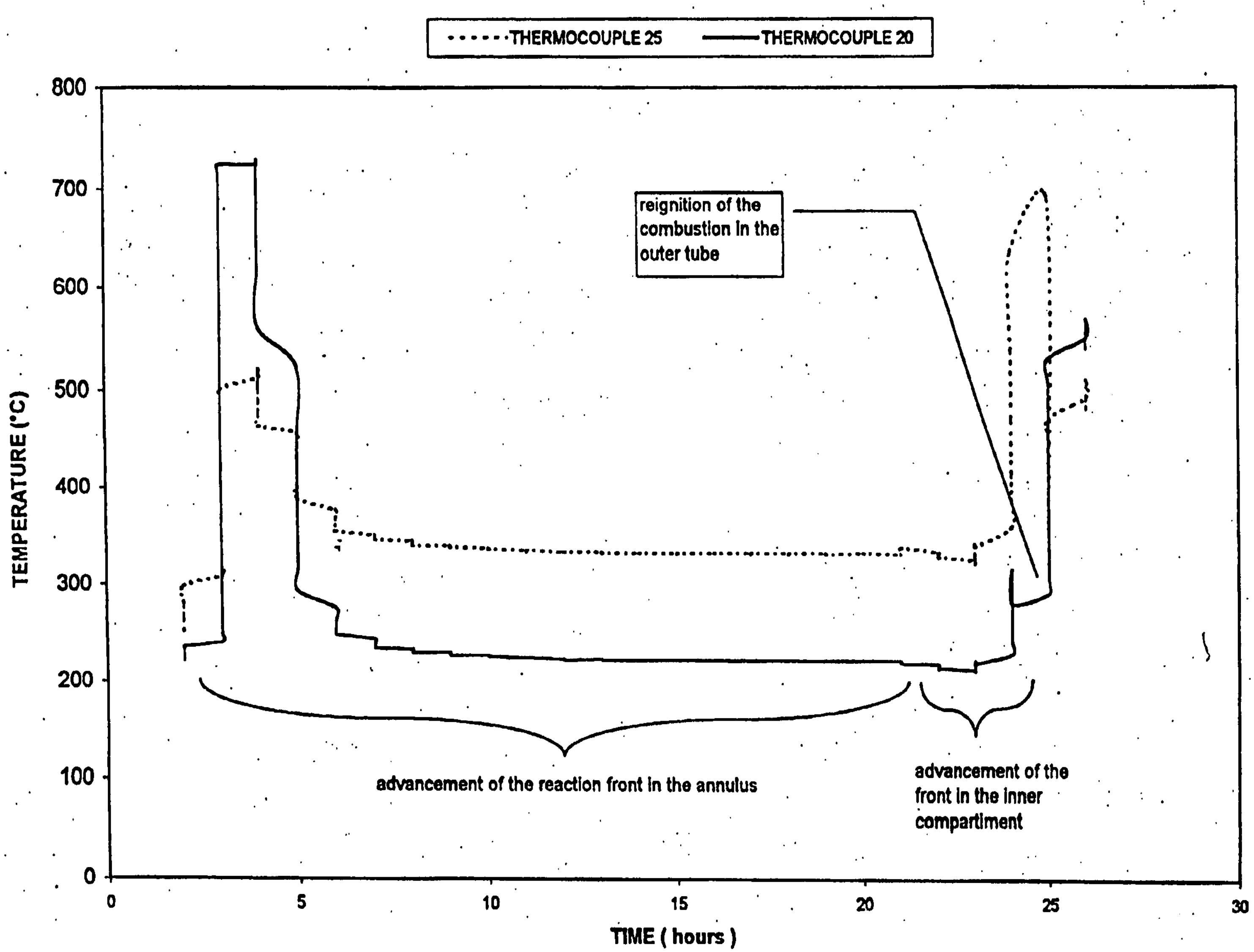

Fig. 13 : Temperature profiles representing autocyclic operation of the experimental self regulating reactor during methan combustion.